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REDSTONE ARSENAL AL RESEARCH DIRECTORATE D W HOWGATE
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I. INTRODUCTION

This report is intended to serve three purposes. The first is to provide some discussion of the structure and functioning of the molecular-electromagnetic correlation formalism (MECF) which is applicable to the study of gases interacting with the electromagnetic field. The second is to present an analysis performed, by the author, during FY 1986 in the application of this formalism to the study of pressure broadening in gases. The third is to briefly summarize past accomplishments and some possible future applications of the approach. It is hoped that the reader will gain some understanding of the nature and scope of this rigorous and comprehensive approach to the description of gas/electromagnetic-field interactions¹.

Because application of the general theory to pressure broadening in gases is only in its initial development state, the analyses presented in Section III is only one milestone in the study of the inclusion of intermolecular interactions due to Coulomb forces in the physical description. The analysis presented is qualitative in nature, and develops the techniques and data needed to evaluate the perturbations induced by increasing the density and/or pressure of the gas in the sample volume considered. Because of its simplicity, the hydrogen atom has been chosen as the "molecular" species used in this initial investigation. It is hoped that many of the qualitative and semiquantitative features of the phenomena of interest can be determined by the use of this "test molecule."

Finally, as a brief indication of the comprehensiveness of the MECF formalism, Section IV outlines several past applications of the method and points out some potential areas for future applications; these latter areas range from a study of the dynamics of the free-electron laser to consideration of adverse-condition imaging in fog or mist. It is hoped that this discussion will stimulate interest in the capabilities of the MECF approach.

¹C. A. Coulter and D. W. Howgate, Physical Review A 31, 843 (1985).

II. BACKGROUND

The MECF formalism is a comprehensive and rigorous method for describing the interaction of a molecular gas with the electromagnetic field. It contains only the very mild physical approximations (see paragraph A below). Up to this point the MECF description has been used in a form that applies to a monomolecular gas, but extension to a mixed gas is possible. The derivation of the MECF description requires the use of quantum field theory. A key step in the development of the theory is the introduction of an operator $P_k(\underline{x}, t)$ that can be thought of as the annihilation operator for a molecule with internal state k and center of mass \underline{x} . However, the MECF formalism itself is expressed solely in terms of non-operator expressions. It contains the energy eigenvalues for the molecular species of interest and also certain functions that are calculated from the wave functions for the molecule. The most important of the functions determined by the wave functions are those shown in paragraph B below. The most important of the dependent variables are discussed in paragraph C.

A. Physical Approximations

Physical approximations contained in the MECF description of the electromagnetic interactions of a molecular gas are:

- Nuclei are treated as elementary particles.
- Nuclei and electrons are treated nonrelativistically.
- The effects of molecular collisions with the walls of the container on the internal molecular states are neglected.

B. Molecular Wave Functions

The most important functions, appearing in the MECF formalism, are determined from the molecular wave functions. Definitions of these functions are given in the following expressions. In these expressions $S_{kl}(\underline{x})$ is a function that specifies the most important part of the coupling of the molecules to the electromagnetic field, $V_{kl|l'k'}(\underline{x})$ is a function that describes the intermolecular interaction, \underline{z} is used to represent the complete set of internal coordinates for the molecule, and $u_k(\underline{z})$ is the wave function for the internal molecular state with index k .

$$S_{kl}(\underline{x}) = \sum_{ij} (\hbar a_j i q_i / 2 i m_i) \int \delta(\underline{x} - \underline{y}_1) [u_k^*(\underline{z}) \nabla_j u_l(\underline{z}) - (\nabla_j u_k^*(\underline{z})) u_l(\underline{z})] \prod_j d^3 z_j \quad (1)$$

and

$$V_{kl|l'k'}(\underline{x}) = \sum_{ii'} q_i q_{i'} \int \frac{[u_k^*(\underline{z}) u_{k'}^*(\underline{z}') u_{l'}(\underline{z}') u_l(\underline{z}) / |\underline{x} - (\underline{y} - \underline{y}')|]}{\prod_j d^3 z_j \prod_{j'} d^3 z_{j'}} \quad (2)$$

Here \underline{x} is the molecular center of mass, \underline{y}_i is the displacement of the i -th particle from the center of mass, a_{ij} is the matrix that transforms laboratory coordinates to internal molecular coordinates, and q_i and m_i are the mass and charge, respectively, of the i -th particle in the molecule.

C. Dependent Variables

The dependent variables in the MECF formalism are a set of correlation functions for the molecules and for the electromagnetic field. The most important of these are the low-order MECFs expressed as expectation values of the electromagnetic vector potential $\underline{A}(\underline{x}, t)$ and the molecular annihilation operator $P_k(\underline{x}, t)$ and its adjoint. V is the volume of the gas, and C_N is a constant dependent on the number N of molecules in the volume and on the number of particles of each species in a molecule. An expression for the value of C_N is given in the paper referenced in Section I.

$$\underline{A}(\underline{x}, t) = \langle \underline{A}(\underline{x}, t) \rangle, P_{k1}(\underline{x}_1, \underline{x}_2, t) = VC_N \langle P_k^+(\underline{x}_1, t) P_1(\underline{x}_2, t) \rangle,$$

$$F^{ab}(\underline{x}_1, \underline{x}_2, t) = \langle A^a(\underline{x}_1, t) A^b(\underline{x}_2, t) \rangle, H^{ab}(\underline{x}_1, \underline{x}_2, t) = c^{-2} \langle \partial_t A^a(\underline{x}_1, t) \partial_t A^b(\underline{x}_2, t) \rangle,$$

$$G^{ab}(\underline{x}_1, \underline{x}_2, t) = (2c)^{-1} \langle A^a(\underline{x}_1, t) \partial_t A^b(\underline{x}_2, t) + \partial_t A^b(\underline{x}_2, t) A^a(\underline{x}_1, t) \rangle,$$

$$Q_{k1}(\underline{x}, \underline{x}_1, \underline{x}_2, t) = VC_N \langle \underline{A}(\underline{x}, t) P_k^+(\underline{x}_1, t) P_1(\underline{x}_2, t) \rangle,$$

$$P_{klmn}(\underline{x}_1, \underline{x}_2, \underline{x}_3, \underline{x}_4, t) = V^2 C_N C_{N-1} \langle P_k^+(\underline{x}_1, t) P_l^+(\underline{x}_2, t) P_m(\underline{x}_3, t) P_n(\underline{x}_4, t) \rangle,$$

$$K_{k1}^{ab}(\underline{x}_1, \underline{x}_2, \underline{x}_3, \underline{x}_4, t) = VC_N \langle A^a(\underline{x}_1, t) A^b(\underline{x}_2, t) P_k^+(\underline{x}_3, t) P_1(\underline{x}_4, t) \rangle. \quad (3)$$

Calculation of the functions S_{k1} and $V_{k11'k'}$, shown in paragraph B above is a sizeable task even for the case of the hydrogen atom, where the "molecular" wave functions are available in analytic form. Determination of the functions in other cases will be even more demanding, but is not out of the question (at least in approximate form) for some of the other simplest atoms and molecules. Figure 1 gives the status of the "data bank" of these functions as of October 1985. The functions T and U referred to in this figure are required for the description of much weaker interactions of the molecules with the electromagnetic field (involving polarization and \underline{A}^2 effects), and have not yet received much study.

MECF DATA BANK (E, S, T, U, V)

FY 85 - 87

Targeted Species

e - electron
H - hydrogen atom
H2 - hydrogen molecule
He - helium

MECF Parameter Matrix Elements

S - principal field/matter parameter
V - matter/matter parameter
T, U - apply only at very short wave lengths and very high fields

SPECIES/PARAMETER MATRIX					MATTER/MATTER MATRIX (V)				
	e	H	H2	He		e	H	H2	He
E	F	F	F	F	e	0	0	0	0
S	0	P5, F6	I5, I6 P7	I5, I6 P7	H		P5, P6 F7	0	0
T	0	0	0	0	H2			P7	0
U	0	0	0	0	He				P7

Definition of Symbols Utilized (not unique to MECF)

0 - no requirement
F - requirement trivial or available from standard sources
I(N) - requirement, but no input as of end of fiscal year 198N
P(N) - requirement, with partial input as of end of fiscal year 198N
F(N) - requirement, with full input as of end of fiscal year 198N
E - energy of targeted species

Figure 1. Summary of the MECF data bank as of 10 October 1985.

The MECF formalism reduces to the standard semiclassical approximation, if one assumes photon wavelengths are significantly greater than molecular dimensions and low-molecular number densities so that the intermolecular interaction terms can be neglected. In addition, it is necessary to assume that the quantum correlations of the electromagnetic field with the molecules can be neglected, so that approximately

$$Q_{k1}(\underline{x}, \underline{x}_1, \underline{x}_2, t) \approx \underline{A}(\underline{x}, t) P_{k1}(\underline{x}_1, \underline{x}_2, t). \quad (4)$$

With these approximations, one is led to a closed set of equations involving only P_{k1} , \underline{A} , and the functions \underline{S}_{k1} and internal molecular energies E_k .

At higher gas pressures, the effects of the intermolecular interaction term involving the functions V_{klmn} must be included. The corresponding set of equations also contains the new MECF P_{klmn} , and to avoid having to extend the set of MECF equations significantly, it is necessary to approximate this four-point correlation function in terms of the two-point functions P_{k1} . Any such approximation must take into account the properties

$$\sum_k \int P_{knn1}(\underline{x}_1, \underline{x}, \underline{x}, \underline{x}_1, t) d^3x = V P_{k1}(\underline{x}_1, \underline{x}_2, t), \quad (5)$$

$$\sum_k \int P_{kk}(\underline{x}, \underline{x}, t) d^3x = v,$$

and

$$P_{klmn}(\underline{x}_1, \underline{x}_2, \underline{x}_3, \underline{x}_4, t) = \pm P_{k1nm}(\underline{x}_1, \underline{x}_2, \underline{x}_4, \underline{x}_3, t), \quad (6)$$

where the + or - sign is chosen in the last equation for an even or odd number of fermions per molecule, respectively. An approximation for P_{klmn} that nearly satisfies the first and third of these conditions (with an error in satisfying the first condition of relative size equal to the ratio of a molecular volume to the total volume of the gas) is

$$P_{klmn}(\underline{x}_1, \underline{x}_2, \underline{x}_3, \underline{x}_4, t) \approx P_{kn}(\underline{x}_1, \underline{x}_4, t) P_{lm}(\underline{x}_2, \underline{x}_3, t) \\ \pm P_{km}(\underline{x}_1, \underline{x}_3, t) P_{ln}(\underline{x}_2, \underline{x}_4, t). \quad (7)$$

This approximation is employed in the following analysis of intermolecular interaction effects to avoid the introduction of higher order MECFs at this point in the development of the implications of the theory.

Note that the quantities necessary to describe the physical properties of the system can be expressed in terms of the MECFs. For example, the expectation values of the molecular number density, molecular momentum density, Poynting vector, and electric and magnetic energy densities can be written as

$$n(\underline{x}, t) = (N/V) \sum_k P_{kk}(\underline{x}, \underline{x}, t),$$

$$\underline{p}(\underline{x}, t) = (N/V) \sum_k (\hbar/2i) [(\nabla_{\underline{x}} - \nabla_{\underline{x}'}) P_{kk}(\underline{x}', \underline{x}, t)] \big|_{\underline{x}' = \underline{x}},$$

$$\langle S^a(\underline{x}, t) \rangle = c \sum_b [\nabla_{\underline{x}}^b G^{ab}(\underline{x}, \underline{x}', t) - \nabla_{\underline{x}}^a G^{bb}(\underline{x}, \underline{x}', t)] \big|_{\underline{x}' = \underline{x}}, \quad (8)$$

$$\langle 1/2 [E(\underline{x}, t)]^2 \rangle = 1/2 \sum_a H^{aa}(\underline{x}, \underline{x}, t),$$

$$\langle 1/2 [B(\underline{x}, t)]^2 \rangle = 1/2 \sum_{a,b} [\nabla_{\underline{x}}^b \nabla_{\underline{x}}^b, F^{aa}(\underline{x}, \underline{x}', t) - \nabla_{\underline{x}}^b \nabla_{\underline{x}}^a, F^{ab}(\underline{x}, \underline{x}', t)] \big|_{\underline{x}' = \underline{x}}.$$

In deriving and illustrating the properties of the MECF formalism, it is desirable to use a simple "molecular" system for which the calculation of the necessary energy values and wavefunction dependent functions S_{kl} is as simple as possible. The hydrogen atom is, of course, ideally suited to this task. Therefore, it is used as the illustrative system in the succeeding analysis of intermolecular interaction effects.

III. ANALYSIS: PRESSURE BROADENING

The information and physical properties sought in the implementation of this discussion can be directly correlated to an analysis of the current vector $\underline{J}(\underline{x}, t)$ that occurs on the right side of the wave equation

$$(\nabla^2 - c^{-2} \partial_t^2) \underline{A}(\underline{x}, t) = -(kc)^{-1} (N/V) \underline{J}(\underline{x}, t), \quad (9)$$

where N/V is the matter density parameter and the initial applied electromagnetic field is assumed to have a plane wave representation of single wave vector $\underline{k} = k\hat{k}$. Since utilization of the hydrogen atom was chosen as the "molecule," the three transitions can now be chosen that will produce the Lyman- α line as the input. In the MECF formalism, these three transitions produce field/matter coefficient vectors $\underline{S}_{lm}^T(\underline{x})$ that lie along the z , x , and y axes, respectively. However, if the direction of propagation of the initial field \hat{k} is chosen to be the z -direction and its vector amplitude A to lie along the x -direction, then \underline{J} will also lie along this direction and now the only concern is with that upper level (labeled 2) which supports an \underline{S}_{lm}^T along the x -direction. In this reduced space one can now express $\underline{J}(\underline{x}, t)$ as

$$\underline{J}(\underline{x}, t) = 2k^3 c \operatorname{Re}[X(t) \underline{A}(\underline{x}, t)], \quad X(t) = (\hbar \Delta)^{-1} (kc)^{-2} S(\underline{k})^2 P_{12}^\alpha(0, t), \quad (10)$$

where

$$S(\underline{k}) = i^{-1} \int d^3x \exp(i\underline{k} \cdot \underline{x}) S_{12}^T(\underline{x}), \quad P_{12}(\underline{R}, \underline{r}, t) = -i(\hbar \Delta c)^{-1} S(\underline{k}) A(\underline{R}, t) P_{12}^\alpha(\underline{r}, t). \quad (11)$$

In arriving at this representation, expression of the principal molecular correlation function as $P_{kl}(\underline{R}, \underline{r}, t)$ was chosen in terms of the coordinates $\underline{R} = (\underline{x}_1 + \underline{x}_2)/2$, $\underline{r} = \underline{x}_1 - \underline{x}_2$. The equation of motion for $P_{12}(\underline{R}, \underline{r}, t)$ has been effectively linearized, by assuming that the diagonal elements $P_{mm}(\underline{R}, \underline{r}, t)$ which appear on the right hand side can be approximated by time independent representations $g_m P^\alpha(\underline{r})$ which are also independent of the global variable \underline{R} . The parameters α and Δ remain to be chosen, whereas the function $X(t)$ is assumed to exhibit the form of a volume susceptibility.

Operating from the left, on the off-diagonal element $P_{12}(\underline{R}, \underline{r}, t)$ one has

$$O'(\underline{R}, \underline{r}, t) = i\partial_t - (\hbar/M)\nabla_{\underline{R}} \cdot \nabla_{\underline{r}} - (E'_2 - E'_1)/\hbar, \quad (12)$$

and only the principal field/matter interaction terms

$$i(c\hbar)^{-1}S(\underline{k}) [g_1 A(\underline{R}-\underline{r}/2, t) - g_2 A(\underline{R}+\underline{r}/2, t)] P^\alpha(\underline{r}),$$

$$A(\underline{x}, t) = A \exp[i(\underline{k} \cdot \underline{x} - kct)] \quad (13)$$

with the Coulomb matter/matter terms

$$(+)(N/V)\hbar^{-1} \int d^3x V_{1122}(\underline{x}) [g_1 P_{12}(\underline{R}-\underline{x}/2, \underline{r}+\underline{x}, t) - g_2 P_{12}(\underline{R}+\underline{x}/2, \underline{r}+\underline{x}, t)] P^\alpha(-\underline{x}) \quad (14)$$

that appear in the MECF formalism will be evaluated on the right-hand side of the $P_{12}(\underline{R}, \underline{r}, t)$ equation of motion. The energies are represented as E'_m in order to imply that they may contain additional elements brought over from the right-hand side of the equation. The matter/matter interaction elements have the following symmetry properties: $V_{imnj}(\underline{x}) = V_{jmni}(\underline{x}) = V_{inmj}(\underline{x}) = V_{mijn}(-\underline{x})$. The intent here is to emphasize the qualitative nature of the analysis and to minimize detail whenever possible. With this in mind, the consideration of terms on the right-hand side containing only diagonal elements, which are independent of the existence of an applied field and introduce corrections to $P_{mm}(\underline{R}, \underline{r}, t)$ of order $(N/V)^2$, is left to the Appendix. The above treatment is consistent with general intent to depict the impact of the Coulomb interaction as an expansion in powers of the molecular density parameter (N/V) or its equivalent.

Now the parameters α and Δ can be developed according to the specifications

$$P^\alpha(\underline{r}) = \exp[-(\alpha r)^2/2], \quad \alpha = (Mk_B T)^{1/2}/\hbar,$$

$$V_{1122}(\underline{x}) = \hbar V \Delta' \int d^3k V_{1122}(\underline{k}), \quad \Delta = (2\pi^2 \hbar V)^{-1} N (q_e a_e)^2, \quad \Delta' = \Delta/N, \quad (15)$$

where T is the temperature, M is the hydrogen atom mass, and q_e and a_e are the electron charge and first Bohr radius, respectively. In so doing, $V_{1122}(\underline{k})$ becomes a dimensionless quantity and the parameters α and Δ have the dimensions of inverse length and inverse time, respectively. The choice of a simple Gaussian for $P^\alpha(\underline{r})$ is a natural consequence of the requirements for the noninteracting gas to be initially ($t=0$) in thermal equilibrium. It further satisfies the additional conditions relevant to including a Coulomb interaction under a perturbation expansion in powers of Δ or N (see Appendix).

Now, for convenience, formally remove α by transforming to a reduced dimensionless representation by way of the relations

$$\begin{aligned} \hbar^{-1}(Mk_B T)^{1/2} \underline{r} \rightarrow \underline{r}, \quad \hbar(Mk_B T)^{-1/2} \underline{s} \rightarrow \underline{s}, \quad (k_B T/M)^{1/2} k t \rightarrow t, \\ (k_B T/M)^{-1/2} k^{-1} \delta \rightarrow \delta, \\ (k_B T/M)^{-1/2} k^{-1} \Delta \rightarrow \Delta, \quad P^\alpha(\underline{r}) \rightarrow P(\underline{r}), \quad P_{12}^\alpha(\underline{r}, t) \rightarrow P_{12}(\underline{r}, t), \quad \underline{t} = t\hat{k}, \quad \underline{\delta} = \delta\hat{k}, \end{aligned} \quad (16)$$

where \underline{s} is an arbitrary wave vector and one is now normalized relative to the influence of thermal ($k_B T$) and impulse ($\hbar k^2/M$) energy. It is this representation, by way of $P_{12}(\underline{r}, t)$, that will provide detailed information of such physical properties as phase and line shape.

Looking at the equation of motion for $P_{12}(\underline{r}, t)$, one sees that

$$O(\underline{r}, t) P_{12}(\underline{r}, t) = R_{12}(\underline{r}, t), \quad O(\underline{r}, t) = \delta/\Delta + i\Delta^{-1}(\partial_t - \hat{k} \cdot \nabla_{\underline{r}}), \quad (17)$$

where $R_{12}(\underline{r}, t)$ is what remains on the right-hand side of the equation after the \underline{r} dependence has been factored out, and δ is just the initial detuning parameter that arises from expression (12) when taking the derivative of $A(\underline{r}, t)$ with respect to time and subtracting the energy level difference. The inverse of $O(\underline{r}, t)$ can be performed through the use of the appropriate Green's function $G_{12}(\underline{x}; t)$, and the relations

$$\begin{aligned} P_{12}(\underline{r}, t) = -i\Delta \int_0^t dt' D(\underline{r}, t-t') R_{12}(\underline{r}, t'), \\ D(\underline{x}, t) = i \int d^3x' G_{12}(\underline{x}-\underline{x}'; t), \end{aligned} \quad (18)$$

a representation in which $D(\underline{x}, t)$ is introduced first, leaving the final integration(s) until later. To properly exploit $D(\underline{x}, t)$, it is convenient at this time to introduce the function

$$Q(\underline{a}; \underline{b}) = \exp[-b^2/2 + i\underline{a} \cdot \underline{b}], \quad Q(\underline{a}+i\underline{x}; \underline{t}) Q(\underline{b}; \underline{x}) = Q(\underline{a}; \underline{t}) Q(\underline{b}+i\underline{t}; \underline{x}), \quad (19)$$

where \underline{a} and \underline{b} are two arbitrary vectors, along with its appropriate integral representation (transform)

$$U(\underline{s}, \underline{x}) = \pi^{-3/2} v^{-1} \int d^3x' Q(\underline{s}; \underline{x}-\underline{x}'). \quad (20)$$

The following important relations governing the application of both $D(\underline{x}, t)$ and $U(\underline{s}, \underline{x})$ can now be developed:

$$\begin{aligned}
 D(\underline{x}, t) Q(\underline{\mu s}; \underline{\mu}^{-1} \underline{x}) &= Q(\underline{\mu}(\underline{s} + \underline{\delta}); \underline{\mu}^{-1} \underline{t}) Q(\underline{\mu s} + \underline{\mu}^{-1} \underline{t}; \underline{\mu}^{-1} \underline{x}) \\
 &= \exp(-i \underline{\delta} \cdot \underline{x}) Q(\underline{\mu}(\underline{s} + \underline{\delta}); \underline{\mu}^{-1}(\underline{t} + \underline{x})), \\
 U(\underline{\mu s}, \underline{\mu}^{-1} \underline{x}) D(\underline{x}, t) Q(\underline{\mu s}'; \underline{\mu}^{-1} \underline{x}) &= V(\underline{\mu})^{-1} \exp[-i \underline{\delta} \cdot \underline{x} - 1/2(\underline{\mu}' s^-)^2] \\
 &\quad \cdot Q(\underline{\mu}'(\underline{s}^+ + \underline{\delta}); (\underline{\mu}')^{-1}(\underline{t} + \underline{x})), \\
 D(\underline{x}, t) U(\underline{\mu s}, \underline{\mu}^{-1} \underline{x}) D(\underline{x}, t') Q(\underline{\mu s}'; \underline{\mu}^{-1} \underline{x}) &= V(\underline{\mu})^{-1} \exp[-i \underline{\delta} \cdot \underline{x} - 1/2(\underline{\mu}' s^-)^2] \\
 &\quad \cdot Q(\underline{\mu}'(\underline{s}^+ + \underline{\delta}); (\underline{\mu}')^{-1}(\underline{t} + \underline{t}' + \underline{x})),
 \end{aligned} \tag{21}$$

where $\underline{s}^+ = (\underline{s} + \underline{s}')/2$, and $\underline{\mu}' = 2^{1/2} \underline{\mu}$ is an arbitrary scaling parameter.

After inspecting equations (18), (19), and (21), it was concluded that as a correlation function $P_{12}(\underline{r}, t)$ is important, primarily, within a radius $r < 1$. Also, that $P_{12}(\underline{r}, t)$ and $R_{12}(\underline{r}, t)$ change rapidly during a brief initial transient period $t < 1$, and then approach values $P_{12}(\underline{r})$ and $R_{12}(\underline{r})$ that are basically independent of time. Thus, for times $t \gg 1$, which are of primary concern, one can write

$$P_{12}(\underline{r}) = -i \Delta \int_0^\infty dt D(\underline{r}, t) R_{12}(\underline{r}), \tag{22}$$

where $R_{12}(\underline{r})$ can be expressed simply in dimensionless representation as

$$\begin{aligned}
 R_{12}(\underline{r}) &= -[g_1 Q(-\underline{k}/2; \underline{r}) - g_2 Q(\underline{k}/2; \underline{r})] \\
 &\quad (\mp) i \pi^{3/2} V \int d^3 k' V_{1122}(\underline{k}') [g_1 U(\underline{k}' + \underline{k}/2, \underline{r}) - g_2 U(\underline{k}' - \underline{k}/2, \underline{r})] \\
 &\quad \cdot \Delta \int_0^\infty dt D(\underline{r}, t) R_{12}(\underline{r}),
 \end{aligned} \tag{23}$$

with the aid of the expressions in (21). Using relations in (21), (22), and (23), it is now easy to write $P_{12}(\underline{r})$ as

$$\begin{aligned}
 P_{12}(\underline{r}) &= i \Delta \exp(-i \underline{\delta} \cdot \underline{r}) \int_0^\infty dt [g_1 Q(-\underline{k}/2 + \underline{\delta}; \underline{t} + \underline{r}) - g_2 Q(\underline{k}/2 + \underline{\delta}; \underline{t} + \underline{r})] \\
 &\quad (\pm) \pi^{3/2} \Delta^2 \exp(-i \underline{\delta} \cdot \underline{r}) \int d^3 k' V_{1122}(\underline{k}') \int_0^\infty dt \int_0^\infty dt' \\
 &\quad \cdot [g_1^2 \exp(-1/4(\underline{k} + \underline{k}')^2) Q(2^{1/2}(\underline{k}'/2 + \underline{\delta}); 2^{-1/2}(\underline{t} + \underline{t}' + \underline{r})) \\
 &\quad - g_1 g_2 \exp(-1/4 \underline{k}'^2) Q(2^{1/2}((\underline{k}' + \underline{k})/2 + \underline{\delta}); 2^{-1/2}(\underline{t}, \underline{t}' + \underline{r}))],
 \end{aligned} \tag{24}$$

where two additional terms involving $g_1 g_2$ and g_2^2 have been omitted to minimize the amount of detail displayed.

It is proper here to think of $Q(\underline{a};\underline{b})$ as one of the primary building blocks developed by this representation of the general formalism. As a product of an oscillatory element $\exp(i\underline{a}.\underline{b})$ and a Gaussian damping term $\exp(-b^2/2)$, its integration over time allows the effect of damping to smooth out the general δ -character produced by the direct time averaging of oscillatory motion. Furthermore, as far as the important Coulomb interaction element V_{1122} and its series expansion terms of arbitrary order $(+)^n$ are concerned, Q (apart from a simple multiplier $\exp(-i\underline{\delta}.\underline{r})$) uniquely displays the functional variation of both \underline{r} and t . For example, for terms of order $(+)^{n-1}$ in the V_{1122} expansion, where the operator U has been applied $n-1$ times, one finds Q displayed as follows:

$$Y_n(\underline{\Omega}, \underline{r}) \exp(-\mu_n^2 \Omega_k^2 / 2) = \int_0^\infty dt_1 \dots \int_0^\infty dt_n Q(\mu_n \underline{\Omega}; (\mu_n)^{-1}(\underline{t}^* + \underline{r})), \quad (25)$$

where $\mu_n = 2^{n/2-1/2}$, $\underline{t}^* = \underline{t}_1 + \dots + \underline{t}_n$, and $\underline{\Omega}$ is an arbitrary vector involving $\underline{\delta}$ and elements of both \underline{k} and the Coulomb interaction wave vector \underline{k}' . This display is a transformation from the basic Q representation into a format where a single Gaussian is weighted by the defined function $Y_n(\underline{\Omega}, \underline{r})$. This function can, in turn, be displayed by using the complex plane to unwind the multiple integration over time, as

$$Y_n(\underline{\Omega}, 0) = \sum_{m=0}^{n-1} (i^m/m!) (\mu_n^2 \Omega_k)^m Y_{n-m}(0, 0) + (i^n/n!) (\mu_n^2 \Omega_k)^n R_n(\Omega_k), \quad (26)$$

$$R_n(\Omega_k) = R_n(-\Omega_k) = (n!/\Omega_k^n) \int_0^{\Omega_k} dz^1 \dots \int_0^z dz^{n-1} \exp[(\mu_n z^n)^2 / 2], \quad (27)$$

where $R_n(\Omega_k)$ is the remainder function for the truncated series expansion and Ω_k , introduced in expressions (25), (26), and (27), is just the projection of the vector $\underline{\Omega}$ along the direction of propagation \underline{k} . The elements $Y_n(0, 0)$ form a series of converging constant terms $Y_1 = (\pi/2)^{1/2}$, $Y_2 = 1$, etc. As correlation is of secondary interest only in this discussion, \underline{r} has been set to equal 0 in developing expressions (26) and (27), for the sake of simplicity of representation. For $\Omega_k^2 < 1$, $R(\Omega_k)$ can be expressed in a Maclaurin series expansion as

$$R_n(\Omega_k) = 1 + \sum_{m=1}^{\infty} (\mu_n^2)^m (2(m-1)+1) \dots (2 \cdot 0 + 1) (n!/(n+2m)!)(\Omega_k)^{2m}, \quad (28)$$

providing a series representation of infinite extent for $Y_n(\underline{\Omega}, 0)$. One can also define the generalized moments

$$M_n^P = \int_{-\infty}^{\infty} d\Omega_k (\Omega_k)^P \exp(-\mu_n^2 \Omega_k^2 / 2) Y_n(\underline{\Omega}, 0) = (-i)^P \int_0^\infty dt_1 \dots \int_0^\infty dt_n \delta^n P(\underline{t}^*) \exp(-(\underline{t}^*/\mu_n)^2 / 2), \quad (29)$$

where $\delta^P(z) = (d^P/dz^P) \delta(z)$, to provide an additional tool for analysis.

The final push comes when one attempts to incorporate expressions such as (25) (or those of order n) into their total environment, which may, in turn, involve additional products of Gaussian elements containing $\underline{\delta}$, \underline{k} , and $\underline{k}' + \underline{k}_1 \dots \underline{k}_{n-1}$ generated by expressions such as those shown in (21), and the Coulomb weighting function

$$\int d\underline{k}_1 V_{1122}(\underline{k}_1) \dots \int d\underline{k}_{n-1} V_{1122}(\underline{k}_{n-1}), \quad (30)$$

which now introduces the impact of the particular molecule involved. An attempt was made to structure this analysis so that this task was put off as long as possible - thus, optimizing the generality of the approach. Although expression (24) depicts Q and its general environment at first and second order in n only, it is basically illustrative of the principal action and processes that are involved. Setting $\underline{r} = 0$ and inserting relation (25), expression (24) can be restated simply as

$$\begin{aligned} P_{12}(0) = & i\Delta[g_1 Y_1(\underline{\delta}-\underline{k}/2, 0) \exp(-(\underline{\delta}-\underline{k}/2)^2/2) - g_2 Y_1(\underline{\delta}+\underline{k}/2, 0) \exp(-(\underline{\delta}+\underline{k}/2)^2/2)] \\ & (+) \pi^{3/2} \Delta^2 \int d^3 \underline{k}' V_{1122}(\underline{k}') \exp[-1/4 \underline{k}'^2 - 1/2(\underline{k}' + \underline{\delta} + \underline{k}/2)^2] \\ & \cdot [g_1^2 Y_2(\underline{\delta}+\underline{k}'/2, 0) \exp(-(\underline{\delta}-\underline{k}/2)^2/2) \\ & - g_1 g_2 Y_2(\underline{\delta}+\underline{k}'/2 + \underline{k}/2, 0) \exp(-(\underline{\delta}+\underline{k}/2)^2/2)], \end{aligned} \quad (31)$$

an expression where the (+) terms display the Gaussian product under the influence of the Coulomb interaction weighted by the product $V_{1122}Y_2$ and averaged over \underline{k}' . The wave vector \underline{k} , where it appears as an impulse correction due to photon emission or absorption (as in the first term of (31)), should have little effect on the impact of pressure broadening and related phenomena at wavelengths far exceeding the Bohr radius of the hydrogen atom and at finite temperatures. It could, in fact, be omitted from expression (31) as a further simplification. The variables k'_1 and k'_2 that appear in expression (31) refer to the components of the vector \underline{k}' along the \hat{k} (or z) direction and on the x - y plane that is normal to it, respectively.

This discussion ends by ignoring the initial population in the upper level (set $g_2 = 0$) and any simple impulse corrections (set $\underline{k} = 0$, except where it occurs inside of $\underline{\delta}$). Now, the ratio $I(\underline{\delta})$ can be formed, using expression (31), of the term of order Δ^2 to the term of order Δ , such that

$$\begin{aligned} I(\underline{\delta}) = & (\mp) i(7\pi/9) \Delta [1 + 1/(2\pi)]^{1/2} R_1(\underline{\delta})^{-1} \int d^3 \underline{k}' \exp[-1/4(\underline{k}'^2_x + \underline{k}'^2_y) - 1/2(\underline{\delta} + \underline{k}'_z)^2] \\ & \cdot \underline{k}'^2_x \underline{k}'^2_y - 2[1 + (2/3)^2 (\underline{\delta} \cdot \underline{k}')^2]^{-6} [1 + 1/(2\pi)]^{1/2} (\underline{\delta} + \underline{k}'_z/2) \\ & - 2(\underline{\delta} + \underline{k}'_z/2)^2 R_2(\underline{\delta} + \underline{k}'_z/2)], \end{aligned} \quad (32)$$

where k'_x , k'_y , and k'_z are the components of \underline{k}' , along the x, y, and z axes, respectively, and a_e is the transformed (dimensionless) value of the hydrogen Bohr radius a_0 . As a limiting approximation, the remainder function $R_n(z)$ will satisfy the relation

$$R_n(z) < n2^{-n/2} \pi^{1/2} / \text{Abs}(z). \quad (33)$$

Using 0.121 micrometers as the wavelength of the Lyman- α line, one is now in the position to evaluate the amplitude scaling factor Δ as a function of pressure and temperature. With the aid of expressions (15) and (16), Δ may be written in the form

$$\Delta = 3.80 \times 10^4 P/T^{3/2}, \quad (34)$$

where pressure P is in atmospheres and temperature T is in degrees Kelvin. At standard condition ($P=1$, $T=300$), then $\Delta = 0.732$ a value slightly below one. Thus, Δ appears at first glance favorable as an initial choice of a scaling parameter and to support the expansion of the Coulomb interaction as a perturbation series involving this parameter. This completes the initial analysis where certain of the (+) terms arising out of the Coulomb interaction have been developed within the confines of the general formalism. This analysis indicates that procedures can be developed for measuring the effectiveness of the general formalism in meeting most requirements in physical applications such as pressure broadening, etc. The further development of such procedures will be a basic part of the next phase of this study.

IV. OTHER APPLICATIONS AND ACCOMPLISHMENTS

As an integral part of prior development of the MECF approach, the following applications have been briefly explored:

1. At the lowest (semiclassical/low density) level of approximation, the impact of gas dynamics on the stimulated emission from an uncorrelated initial excited state in a gas was developed through a calculation of the transition probability to the ground state. Further complexity was introduced into the final arrangement of photon, thermal, and molecular parameters by the addition of a sound wave. In particular, the precise detuning needed to produce a maximum response of stimulated emission directly reflects the impact of the added sound wave on the radiation process. Also, a three-level model was discussed with the procedures for consideration of such related processes as stimulated Raman scattering¹.

2. To explore the potential of a traveling current-initiated (in the container walls) induced-dipole laser in molecular hydrogen gas, where the expectation value of the vector potential \underline{A} is always zero, the (purely quantum) level of approximation to the MECF formalism was partially developed. It yielded a rigorous fully dimensional traveling wave description for both spontaneous emission and induced-dipole emission in a gas. Specifically, the ratio of the rate of population change at time t in the induced dipole case to that in the case where a direct non-induced transition can occur was directly exhibited. In this purely quantum example, \underline{A} can no longer characterize the electromagnetic field. It is actually the source correlation functions which couple the molecule to \underline{A} and its partial derivative that determine the Poynting vector and so characterize the field. (Results have not been published.)

3. To review the process of excitation of a molecular system (hydrogen) by electron impact (as in a discharge or e-beam), the MECF formalism was displayed for these two mutually interacting species. The impact of pressure broadening was briefly considered and a group of new matrix elements specifically involved in the excitation process were calculated for a subset of the quantum states of molecular hydrogen. (Results have not been published.)

Because of its inherent ability to separate internal structure (symbolized by the wave function u_k or just k) from center of mass motion (symbolized by the coordinate \underline{x}), the MECF approach is particularly applicable to processes such as Raman and Brillouin scattering which exhibit contrasting profiles relative to these specific properties. Another area of direct potential relevance for the MECF approach is the investigation of Rydberg-state interactions. Here, one has the specific ability to handle a complex gaseous mix involving low lying states, highly excited states, ions, electrons, a buffer gas, and (of course) the electromagnetic field. Also, one can present numerous other applications, both standard and more exotic, such as the impact of light-induced drift and light pressure sound waves in a gas, etc. The specific introduction of long-range interaction character, introduced through taking the transverse part of $S_{k1}(\underline{x})$, with application to individual atoms and molecules would likewise represent a potential area of associated interest.

¹Charles Wang, Ed., "Gas Effects in Stimulated Emission," in Lasers in Fluid Mechanics and Plasmadynamics, AIAA, New York, 1983.

Finally, a few potential areas of application that extend the limits of the formalism are mentioned. One, previously alluded to, is an in-depth consideration of the interaction of electromagnetic waves with a fog or mist. Here, one has increased the fundamental size of the working unit from that of a molecule to that of a group of molecules, etc., that can be described collectively as having both a center of mass and an internal character or structure analogous to that of a large molecule. Increasing gaseous pressure and density, on the other hand, eventually moves the MECF approach toward a consideration of transitions in phase. Application here becomes limited when it is no longer possible to separate out the individual molecule. Applications involving a consideration of lower-order relativistic effects or corrections appear to be more attractive where one is not, as yet, forced to move from an initial Hamiltonian form to a consideration of alternative Lagrangian formulations. A major application here would be a rigorous characterization and treatment of the free-electron laser, where the basic unit is now the electron and relativistic effects have been introduced. Surface shielding effects, involving the interaction of plasmas with radiation or particle beams, represent another area of advanced activity treatable by the MECF approach. Here the specification of adequate boundary conditions would be particularly challenging. Additional applications can also be cited in the general area of collective processes and phenomena.

APPENDIX

An issue that cannot be ignored is the impact of the Coulomb interaction in modifying the diagonal elements of the particle density matrix in the absence of any external or applied electromagnetic field. Any modification of this nature would become directly applicable to our study, once an external field was added to the system. Because this modification first arises at order N^2 , in a perturbation expansion on the Coulomb interaction, it was not directly addressed in the text, which chose to consider only terms involving N^0 and N^1 . However, since the power of N is not an absolute guide to (or measure of) impact on specific physical properties and observables, an analysis that can be used toward the clarification of this important subject is developed in this Appendix.

As a physical example, the hydrogen atom is again chosen, along with its ground state 1, the impact of the three levels 2(j) that produce the Lyman- α line are investigated. For convenience, at this time, off-diagonal correlation functions are expressed in the form $P_{\pm j}^{\pm}(\underline{r}, t) = P_{12(j)}^{\pm}(\underline{r}, t) + P_{2(j)1}^{\pm}(\underline{r}, t)$, which due to the absence of an external field should be independent of the global variable R . Furthermore, if the diagonal elements $P_{mm}(\underline{r}, t)$ when they appear on the right hand side of the equations of motion are allowed to assume the zeroth order form $g_m P^0(\underline{r})$, or a simple Gaussian in \underline{r} , and then transformed to a reduced dimensionless representation (see expressions (16)), the equations of motion for $P_{\pm j}^{\pm}(\underline{r}, t)$ are simply

$$i\partial_t P_{-j}^{-}(\underline{r}, t) - \Omega_{12} P_{+j}^{+}(\underline{r}, t) = 0,$$

$$i\partial_t P_{+j}^{+}(\underline{r}, t) - \Omega_{12} P_{-j}^{-}(\underline{r}, t) = (+)2\pi^{3/2}g_{-}^{-2}\Omega_{12}N \int d^3k v_{12}^j(\underline{k}) Q(-2^{-1/2}\underline{r}; 2^{-1/2}\underline{k}), \quad (A-1)$$

where the dimensionless $v_{12}^j(\underline{k})$ is just $v_{12}^j(\underline{k}) = (g_{-}\hbar\Omega_{-}V)^{-1} \sum_m g_m v_{m2(j)1}(\underline{k})$ with the summation over all four states, 12 , and $\Omega_{12} = (E_{2(j)} - E_1)/\hbar$, $g_{-} = g_1 - g_2(j)$ where there is no direct dependence on j . In arriving at this simple format, the fact that the integral over all space of $v_{m2(j)1}(\underline{x})$ is zero has been used and higher order terms have been neglected on the right hand side of the equations. Since time t and Ω_{12} , in a sense, act independently in this development where they are directly coupled, they may be considered to be in either the original or the reduced dimensionless form. Also, the function Q , as it appears in expressions (A-1), is just the Q of expression (19).

As a measure of the correction on the diagonal elements $P_{mm}(\underline{r}, t)$ at order N^2 , the quantity $\Delta P(\underline{r}, t) = \Delta P_{11}(\underline{r}, t) - \sum_j \Delta P_{2(j)2(j)}(\underline{r}, t)$ is used, which folds all of the three 2(j) levels into the mix. In this way, one is able to illustrate the more general impact of the Coulomb interaction, not being tied to a single 2(j) level. Using the same approach as was taken in the construction of expressions (A-1), one can now write the equation of motion for $\Delta P(\underline{r}, t)$ as

$$i\partial_t \Delta P(\underline{r}, t) = (+)2g_{-}^{-2}\Omega_{12}N \int d^3k \int d^3x \exp(i\mathbf{k} \cdot \mathbf{x}) P(\underline{x}) \sum_j v_{12}^j(\underline{k}) P_{+j}^{+}(\underline{x} + \underline{r}, t), \quad (A-2)$$

where only P^+ is directly involved. Assuming $\Delta P(\underline{r}, 0) = 0$, expressions (A-1) and (A-2) yield a simple solution with oscillatory time dependence

$$\Delta P(\underline{r}, t) = (32/3^{1/2})(\pi g_-)^3 N^2 (1 - \cos \Omega_{12} t)$$

$$P(\underline{r}') \int d^3 \underline{k} \int d^3 \underline{k}' \sum_j v_{12}^j(3^{1/2} \underline{k}) v_{12}^j(3^{1/2} \underline{k}')$$

$$\cdot \exp[-(k^2 - \underline{k} \cdot \underline{k}' + k'^2)] \exp[-i(\underline{k} + \underline{k}') \cdot \underline{r}'], \quad (A-3)$$

where $\underline{r}' = 3^{-1/2} \underline{r}$ and $P(\underline{r})$ is the initial Gaussian functional form. Although expression (A-3) illustrates the analytic nature of $\Delta P(\underline{r}, t)$, and thus ΔP_{11} and $\Delta P_{2(j)2(j)}$ through the obvious relations $\Delta P_{11} = 1/2 \Delta P$, $\sum_j \Delta P_{2(j)2(j)} = -1/2 \Delta P$, only by inserting the latter in place of $P(\underline{r})$ in expressions such as the transformed version of (13) can one truly evaluate its quantitative significance. It is obvious from (A-3) that $\Delta P(\underline{r}, t)$ is of even parity in \underline{r} and is saved from approaching zero as \underline{r} approaches zero by the cross term $\underline{k} \cdot \underline{k}'$ that couples the \underline{k} and \underline{k}' integrals together. Further details on this subject will be developed at a later time.

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